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U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

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**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. §371**

WEICKM 10

U.S. APPLICATION NO. (If known, see 37 CFR §1.5)

09/831825

INTERNATIONAL APPLICATION NO.

INTERNATIONAL FILING DATE

PCT/EP99/08780

15 NOVEMBER 1999

PRIORITY DATE CLAIMED

16 NOVEMBER 1998

TITLE OF INVENTION

CRYSTALLINE POROUS SOLIDS, PRODUCTION AND USE THEREOF

APPLICANT(S) FOR DO/EO/US


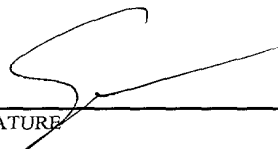
CHANDRA, Amita, et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. §371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. §371.
3. ☐ This express request to begin national examination procedures (35 U.S.C. §371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. §371(b) and PCT Articles 22 and 39(1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. §371(c)(2))
 - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☒ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A translation of the International Application into English (35 U.S.C. §371(c)(2)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. §371(c)(3))
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☒ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☐ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. §371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. §371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. §371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 C.F.R. §§1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 C.F.R. §§3.28 and 3.31 is included.
13. ☒ A **FIRST** preliminary amendment.
 - ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☐ Other items or information:

U.S. APPLICATION NO. 097/831825 INTERNATIONAL APPLICATION NO. PCT/EP99/08780		ATTORNEY'S DOCKET NUMBER WEICKM 10	
17. <input checked="" type="checkbox"/> The following fees are submitted: BASIC NATIONAL FEE (37 CFR §1.492 (a) (1) - (5)): Search Report has been prepared by the EPO or JPO..... \$860.00 International preliminary examination fee paid to USPTO (37 CFR §1.482)..... \$690.00 No international preliminary examination fee paid to USPTO (37 CFR §1.482) but international search fee paid to USPTO (37 CFR §1.445(a)(2))..... \$710.00 Neither international preliminary examination fee (37 CFR §1.482) nor international search fee (37 CFR §1.445(a)(2)) paid to USPTO..... \$1000.00 International preliminary examination fee paid to USPTO (37 CFR §1.482) and all claims satisfied provisions of PCT Article 33(2)-(4)..... \$100.00 ENTER APPROPRIATE BASIC FEE AMOUNT = \$860.00		CALCULATIONS PTO USE ONLY	
Surcharge of \$130.00 for furnishing the oath or declaration later than months from the earliest claimed priority date (37 C.F.R. §1.492(e)). <input type="checkbox"/> 20 <input type="checkbox"/> 30			
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE
Total claims	16 - 20 =	0	x \$ 18.00
Independent claims	1 - 3 =	0	x \$ 80.00
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$ 270.00
TOTAL OF ABOVE CALCULATIONS =			\$860.00
Reduction of 1/2 for filing by small entity, if applicable. A Verified Small Entity Statement must also be filed (Note 37 C.F.R. §§1.9, 1.27, 1.28).			
SUBTOTAL =			\$860.00
Processing fee of \$130.00 for furnishing the English translation later than months from the earliest claimed priority date (37 C.F.R. §1.492(f)). <input type="checkbox"/> 20 <input type="checkbox"/> 30			
TOTAL NATIONAL FEE =			\$860.00
Fee for recording the enclosed assignment (37 C.F.R. §1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 C.F.R. §§3.28, 3.31). \$40.00 per property.			
TOTAL FEES ENCLOSED =			\$860.00
			Amount to be refunded:
			charged:
a. <input checked="" type="checkbox"/> A check in the amount of <u>\$860.00</u> to cover the above fees is enclosed. b. <input type="checkbox"/> Please charge my Deposit Account No. <u>13-3402</u> in the amount of \$_____ to cover the above fees. A duplicate copy of this sheet is enclosed. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>13-3402</u> . A duplicate copy of this sheet is enclosed.			
NOTE: Where an appropriate time limit under 37 C.F.R. §§1.494 or 1.495 has not been met, a petition to revive (37 C.F.R. §1.137(a) or (b)) must be filed and granted to restore the application to pending status.			
SEND ALL CORRESPONDENCE TO. Customer Number 23,599			
 23599 PATENT TRADEMARK OFFICE		SIGNATURE  Anthony J. Zelano NAME	
Filed: 15 MAY 2001 AJZ:kms		27,969 REGISTRATION NUMBER	

IN THE UNITED STATES DESIGNATED/ELECTED OFFICE

International Application No. : PCT/EP99/08780
International Filing Date : 15 NOVEMBER 1999
Priority Date(s) Claimed : 16 NOVEMBER 1998
Applicant(s) (DO/EO/US) : CHANDRA, Arnita, et al.

Title: CRYSTALLINE POROUS SOLIDS, PRODUCTION AND USE THEREOF

PRELIMINARY AMENDMENT

Commissioner for Patents
Washington, D.C. 20231

SIR:

Although the claims of the above application were amended under Article 34 during the International Phase, applicants respectfully request that examination be based on the claims as originally filed and this Preliminary Amendment is based thereon.

Prior to calculating the national fee, and prior to examination in the National Phase of the above-identified International application, please amend as follows:

IN THE CLAIMS:

3. (Amended) The method as claimed in claim 1, characterized in that the fluid mixture has an essentially eutectic composition.
4. (Amended) The method as claimed in claim 1, characterized in that the second phase is removed in step (iii) by means of solvent extraction.
5. (Amended) The method as claimed in claim 1, characterized in that the second phase is a substance which is soluble in aqueous media.
6. (Amended) The method as claimed in claim 1, characterized in that the first phase is a water-insoluble salt.

7. (Amended) The method as claimed in claim 1, characterized in that the second phase is a water-soluble salt which is able to form a eutectic mixture with the first phase.

8. (Amended) The method as claimed in claim 1, characterized in that the first phase comprises AgCl and the second phase comprises an alkali metal halide.

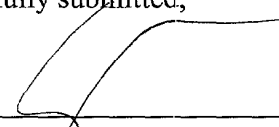
10. (Amended) Porous ion-conducting solid obtainable via a method as claimed in claim 1.

14. (Amended) The use of a solid or an electrochemical cell as claimed in claim 11 as a sensor.

REMARKS

The purpose of this Preliminary Amendment is to eliminate multiple dependent claims in order to avoid the additional fee. Applicants reserve the right to reintroduce claims to canceled combined subject matter.

Respectfully submitted,



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AJZ:jmm

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VERSION WITH MARKINGS TO SHOW CHANGES MADE

Claims 3-8, 10 and 14 have been amended as follows:

3. (Amended) The method as claimed in ~~any one of claims 1 or 2~~, characterized in that the fluid mixture has an essentially eutectic composition.
4. (Amended) The method as claimed in ~~any one of claims 1 to 3~~, characterized in that the second phase is removed in step (iii) by means of solvent extraction.
5. (Amended) The method as claimed in ~~any one of claims 1 to 4~~, characterized in that the second phase is a substance which is soluble in aqueous media.
6. (Amended) The method as claimed in ~~any one of the preceding claims 1~~, characterized in that the first phase is a water-insoluble salt.
7. (Amended) The method as claimed in ~~any one of the preceding claims 1~~, characterized in that the second phase is a water-soluble salt which is able to form a eutectic mixture with the first phase.
8. (Amended) The method as claimed in ~~any one of the preceding claims 1~~, characterized in that the first phase comprises AgCl and the second phase comprises an alkali metal halide.
10. (Amended) Porous ion-conducting solid obtainable via a method as claimed in ~~any one of claims 1 to 9~~.
14. (Amended) The use of a solid or an electrochemical cell as claimed in ~~any one of claims 11 to 13~~ as a sensor.

CRYSTALLINE POROUS SOLIDS, PRODUCTION AND USE THEREOF

Description

- 5 The invention relates to crystalline porous solids, to a method of producing them and to the use of them.

10 Porous solids have attracted increasing interest in years past. They combine the characteristics of a massive solid, such as mechanical strength, with certain application-specific advantages such as e.g. reduced weight, high surface area, in some cases permeability to relatively small molecules or exclusion of relatively large molecules etc. Important fields of application for the porous materials are therefore found in separation technology or catalysis.

15 Previously known porous solids consist of ceramic materials having low conductivity and are produced via precursor methods or decomposition methods. They are therefore of no use or of only limited use as conductive materials (e.g. in sensor technology).

20 Such a method of producing porous solids having low conductivity is described e.g. in DE 41 02 430 A1. According to DE 41 02 430 A1, a fine-pored solid having a high pore volume is produced by a coarsely disperse sedimentable mixture of a liquid phase and solid particles being caused to sediment and the sediment being solidified in the presence of the liquid phase by a chemical reaction between the sediment particles to form a porous body.

25 DE 37 31 649 A1 describes a method of producing open-pored sintered bodies which comprises sintering a sinterable glass powder and an inorganic, soluble salt of defined granularity, whose melting point is above the melting temperature of the sinterable powder, and dissolving away, after cooling, the soluble salt

present in the sintered body. The glass powder used according to DE 37 31 649 A1 is distinguished, in particular, by high sinterability. This method too affords open-pored sintered bodies which consist of
5 glass ceramics and have low conductivity.

There is therefore a need for porous materials having a conductivity which is higher than that of the known porous ceramics.

10

The present invention therefore relates to a method of producing a porous solid, characterized by the steps of:

- 15 (i) preparing a fluid mixture comprising a first phase which includes one or more inorganic ionic components, and at least one second phase, the first phase and the second phase being essentially immiscible in the solid state,
- 20 (ii) cooling the fluid mixture to a temperature below the solidification point in order to form a solid phase mixture comprising at least one first crystalline phase and a second phase, and
- (iii) removing the second phase.

25 The advantages of the method described here consist in the simplicity of the production of porous conductive solids, which involves the preparation of a preferably eutectic mixture of at least two solid phases, at least one soluble and one insoluble phase. The morphology of
30 the phases can be influenced by simple variation of the preparation conditions or of the quenching rate. Removal of the soluble phase results in the formation of an open pore network. Thus, porous conductive, especially ion-conducting, electroceramics are
35 accessible which owing to the microstructure of the eutectic have high mechanical stability. The significance of the method inter alia resides in the fact that - as shown in the examples - the highly

porous materials obtained provide a large contact area and are therefore of importance for the use in sensorics, e.g. in gas sensorics. Equally, the solids according to the invention can also serve as containers
5 for a liquid electrolyte. Owing to interface interactions, the solid can be filled in a simple manner with electrolyte liquid and subsequent bleeding of the latter (phase separation) can be prevented.

10 The porous solid resulting from the method has an essentially open-pored structure and, owing to its crystalline composition has a high conductivity, especially ionic conductivity. The mean pore size depends on the structure of the phase which has been
15 dissolved away and can therefore vary over a wide range. Thus, the pores can e.g. have a size in the order of from about 20 nm to 5 μm in each spatial direction. Anisotropic pore structure can likewise be obtained, e.g. lamellar pore structures, which can have
20 pore sizes of from 2 to 3.5 μm \times 500 nm up to 1.5 μm \times 20 nm to 200 nm. The degree of the porosity (fraction of the pore volume relative to the total volume) depends on the respective fractions of the first and second phases in the fluid mixture and can
25 range from about 10 to 70%, preferably from 20 to 50%.

The fluid mixture prepared in step (i) of the method according to the invention includes at least two phases, which are miscible in the fluid state but not
30 in the solid state. The term "fluid state" refers to a melt or alternatively e.g. a plasma. The first phase includes one or more inorganic ionic components, especially ionic compounds such as salts, for example. Preferred examples of such compounds are
35 water-insoluble salts, e.g. silver halides, especially AgCl.

The second phase comprises a substance which in the solid state is immiscible with the first phase and preferably is essential miscible therewith in the fluid state. Preferably, the second phase used is a water-soluble salt compound which is able to form a eutectic mixture with the first phase. If the first phase is a silver halide, e.g. AgCl, the second phase used can be an alkaline earth metal halide or alkali metal halide, e.g. KCl, RbCl or/and CsCl. Particularly preferred is a eutectic mixture comprising about 70 mol% of AgCl and 30 mol% of KCl.

According to step (ii) the fluid mixture is cooled to a temperature below the solidification point. This affords a solid which contains a phase mixture comprising at least one first crystalline phase and a second, selectively removable phase. It is also possible for further phases to be present, which can be selectively removable soluble phases or/and insoluble phases which remain in the resulting porous solid.

Via the cooling rate it is possible to vary the morphology of the resulting solid. According to one embodiment, cooling takes place under non-segregating conditions (quenching), the cooling rate being sufficiently high to prevent crystal growth and consequently the formation of substantial crystals. In this case, the cooling rate is preferably in the range of from 10 to 50°C/min and above. In other cases, slower cooling is also possible, to enable crystal growth to the extent desired. Thus, slow cooling of a non-eutectic fluid mixture first results in a fluid eutectic composition containing particles of the first or second phase dispersed therein, which then solidifies as the temperature drops below the eutectic temperature. It is thus possible to produce a porous solid which includes two or more pore species which differ in terms of size or/and morphology.

Preferably, the fluid mixture has an essentially eutectic phase composition. Upon cooling of such a mixture, porous solids having a lamellar morphology can be obtained. The composition of the mixture is preferably in the range of ± 10 mol%, especially ± 2.5 mol% of a eutectic mixture.

The removal of the second phase from the solid can be effected, for example, by solvent extraction, if the first phase is insoluble in a particular solvent and the second phase is soluble therein. Preferentially, a second substance is used which is soluble in aqueous media (water, aqueous acid or bases). Where appropriate, however, organic solvents can also be used for the extraction.

As an alternative to a solvent extraction, the second phase can also be removed by other methods (chemical reactions or/and heating).

The invention further relates to a porous ion-conducting solid obtainable via the inventive method.

The porous solid can be employed directly for further use. Alternatively, however, it can also be ground down into smaller particles and be converted into a different shape, e.g. by compression molding. If the solid consists of an ion-conducting material it can be used in an electrochemical cell as an electrolyte, e.g. as a solid electrolyte or as a support for a liquid electrolyte. The electrochemical cell customarily contains at least two electrodes (e.g. measuring electrode and reference electrode) and the electrolyte disposed between the electrodes. The cell can be used as a sensor, e.g. as an amperometric or conductometric sensor for determining physical parameters, e.g. temperature, or chemical parameters, e.g. gaseous

substances such as H_2O , CO_2 and NH_3 . By using the inventive porous solids as electrolytes, a considerable increase in the sensitivity of such sensors is possible. A porous AgCl solid is suitable, in particular, for the determination of NH_3 .

The porous solid is also suitable for other applications (fluid supports, separation techniques, catalysis). For these purposes, the pores of the solid can, if required, be coated with further substances, e.g. metals, metal oxides or even biomolecules.

The invention is explained in more details by the following examples, in conjunction with the appended figures in which:

15

Figure 1a is a scanning electron micrograph of the lamellar structure which was obtained by cooling a fluid mixture of a eutectic composition of AgCl and KCl (30 mol% KCl , 70 mol% AgCl),

20

Figure 1b is a scanning electron micrograph of the porous AgCl solid obtained after the KCl phase had been dissolved away,

25

Figure 2a is a diagram which shows the reversible change in the conductivity in a porous AgCl solid upon switching from an Ar atmosphere to an NH_3 atmosphere and back, and

30

Figure 2b shows the change in the conductivity in a porous AgCl solid in the absence and the presence of a liquid electrolyte (0.5 M and 1 M AgNO_3) as a function of the temperature.

35

Examples

1. Production of a porous AgCl solid

AgCl (70 mol%) and KCl (30 mol%) are heated to 350°C in a preheated furnace. The homogeneous melt is cooled to room temperature by being removed from the furnace.

5 Then the KCl is dissolved away by immersion in distilled water, and the resulting solid is dried in air for 24 h. A porous, mechanically stable solid is obtained. The porosity corresponds to the KCl content.

10 The structure of the solid before and after the extraction of KCl is shown in figures 1a and 1b.

2. Determination of NH₃

15 The porous AgCl solid according to example 1 is ground to a powder which is then compression-molded by uniaxial compression, using a pressure of about 30 kN/cm², to produce pellets having a diameter of 1 cm.

20 A pellet is arranged between two electrodes to produce an NH₃ sensor. Silver paste is used for the electrodes.

Figure 2a shows the change in conductivity of the porous AgCl specimen in the presence of NH₃ and inert gas (argon), respectively. A reproducible and reversible rapid change in conductivity proportional to the NH₃ concentration is observed.

30 3. Liquid-electrolyte support

A porous AgCl solid produced in accordance with example 1 is filled with liquid electrolyte (AgNO₃). Owing to the capillary forces, the liquid electrolyte is readily
35 absorbed by the porous solid and retained therein. Figure 2b shows the change in the conductivity of a porous AgCl solid in the presence and absence of AgNO₃ (0.5 M and 1 M) as a function of the temperature. As

can be gathered from the diagram, the porous AgCl solid is eminently suitable as a support for liquid electrolytes.

Claims

1. A method of producing a porous solid,
characterized by the steps of:
- 5 (i) preparing a fluid mixture comprising a first
phase which includes one or more inorganic
ionic components, and at least one second
phase, the first phase and the second phase
being essentially immiscible in the solid
10 state,
- (ii) cooling the fluid mixture to a temperature
below the solidification point in order to
form a solid phase mixture comprising at
least one first crystalline phase and a
15 second phase, and
- (iii) removing the second phase.
2. The method as claimed in claim 1,
characterized in that
- 20 the cooling is performed under non-segregating
conditions.
3. The method as claimed in any one of claims 1 or 2,
characterized in that
- 25 the fluid mixture has an essentially eutectic
composition.
4. The method as claimed in any one of claims 1 to 3,
characterized in that
- 30 the second phase is removed in step (iii) by means
of solvent extraction.
5. The method as claimed in any one of claims 1 to 4,
characterized in that
- 35 the second phase is a substance which is soluble
in aqueous media.

6. The method as claimed in any one of the preceding claims,
characterized in that
the first phase is a water-insoluble salt.
- 5 7. The method as claimed in any one of the preceding claims,
characterized in that
the second phase is a water-soluble salt which is
10 able to form a eutectic mixture with the first phase.
8. The method as claimed in any one of the preceding claims,
15 characterized in that
the first phase comprises AgCl and the second phase comprises an alkali metal halide.
9. The method as claimed in claim 8,
20 characterized in that
the mixture is formed from 70 mol% of AgCl and 30 mol% of KCl.
10. Porous ion-conducting solid obtainable via a
25 method as claimed in any one of claims 1 to 9.
11. An electrochemical cell which contains as the electrolytes a porous solid as claimed in claim 10.
- 30 12. The electrochemical cell as claimed in claim 11,
characterized in that
the pores of the solid are filled with a fluid.
- 35 13. The electrochemical cell as claimed in claim 12,
characterized in that
the fluid is a liquid electrolyte.

14. The use of a solid or an electrochemical cell as claimed in any one of claims 11 to 13 as a sensor.
15. The use as claimed in claim 14 for the
5 determination of gases.
16. The use of a solid as claimed in claim 10 in separation technology or in catalysis.

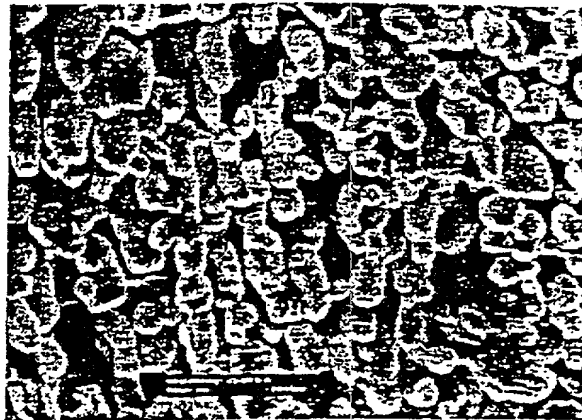
11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 30 31 32 33 34 35 36 37 38 39 40 41 42 43 44 45 46 47 48 49 50 51 52 53 54 55 56 57 58 59 60 61 62 63 64 65 66 67 68 69 70 71 72 73 74 75 76 77 78 79 80 81 82 83 84 85 86 87 88 89 90 91 92 93 94 95 96 97 98 99 100 101 102 103 104 105 106 107 108 109 110 111 112 113 114 115 116 117 118 119 120 121 122 123 124 125 126 127 128 129 130 131 132 133 134 135 136 137 138 139 140 141 142 143 144 145 146 147 148 149 150 151 152 153 154 155 156 157 158 159 160 161 162 163 164 165 166 167 168 169 170 171 172 173 174 175 176 177 178 179 180 181 182 183 184 185 186 187 188 189 190 191 192 193 194 195 196 197 198 199 200 201 202 203 204 205 206 207 208 209 210 211 212 213 214 215 216 217 218 219 220 221 222 223 224 225 226 227 228 229 230 231 232 233 234 235 236 237 238 239 240 241 242 243 244 245 246 247 248 249 250 251 252 253 254 255 256 257 258 259 260 261 262 263 264 265 266 267 268 269 270 271 272 273 274 275 276 277 278 279 280 281 282 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Figure 1a



Figure 1b



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Figure 2a

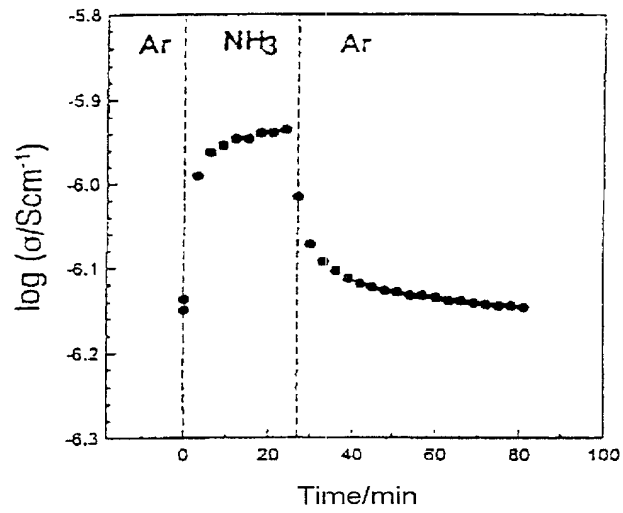
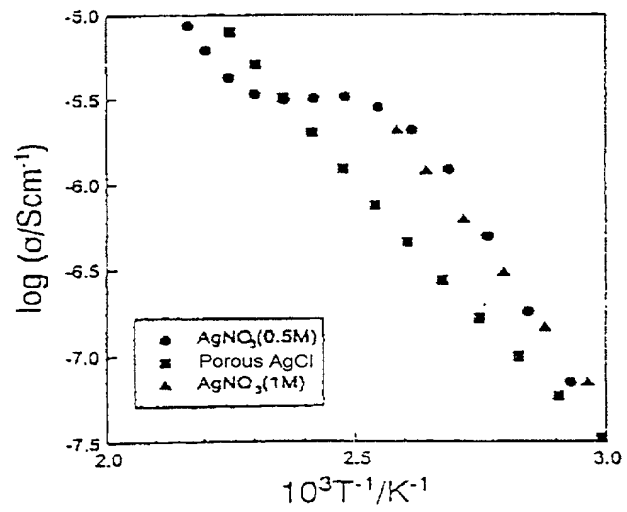


Figure 2b



DECLARATION FOR PATENT APPLICATION

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

CRYSTALLINE POROUS SOLIDS, PRODUCTION AND USE THEREOF

the specification of which

☐ is attached hereto

☒ was filed on 15 NOVEMBER 1999 as United States Application Number or PCT International Application Number PCT/EP99/08780 and (if applicable) was amended on _____

I hereby authorize our attorneys to insert the serial number assigned to this application.

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR §1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

PRIOR FOREIGN/PCT APPLICATION(S) AND ANY PRIORITY CLAIMS UNDER 35 USC §119			
APPLICATION NO.	COUNTRY	DAY/MONTH/YEAR FILED	PRIORITY CLAIMED
198 52 783.7	GERMANY	16 NOVEMBER 1998	YES

I hereby claim the benefit under 35 U.S.C. §119(e) of any United States provisional application(s) listed below.

PROVISIONAL APPLICATION(S) UNDER 35 U.S.C. §119(e)	
APPLICATION NUMBER	FILING DATE

I hereby claim the benefit under 35 U.S.C. §120 of any United States application, or §365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. §112.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR §1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

PRIOR U.S./PCT INTERNATIONAL APPLICATION(S) DESIGNATED FOR BENEFIT UNDER 37 U.S.C. §120		
APPLICATION NO.	FILING DATE	STATUS — PATENTED, PENDING, ABANDONED

I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected herewith: I. William Millen (19,544); John L. White (17,746); Anthony J. Zelano (27,969); Alan E.J. Branigan (20,565); John R. Moses (24,983); Harry B. Shubin (32,004); Brion P. Heaney (32,542); Richard J. Traverso (30,595); John A. Sopp (33,103); Richard M. Lebovitz (37,067); John H. Thomas (33,460); Catherine M. Joyce (40,668); Nancy J. Axelrod (44,014); James T. Moore (35,619); James E. Ruland (37,432); Jennifer J. Branigan (40,921) and Robert E. McCarthy (46,044)

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PATENT TRADEMARK OFFICE

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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☐ Additional joint inventors are named on separately numbered sheets attached hereto.